1. Brief Review of the Prior Art

Haruta '394 discloses a method for the production of ultra-fine gold particles on a metal oxide support, such as titania, by combining in solution a gold compound, a metal salt, and a carboxylic acid at controlled pH to induce co-precipitation of gold hydroxide onto the metal oxide, and then heating the precipitate to obtain ultra-fine gold particles on the metal oxide support. Haruta '394 is silent with respect to impregnation. The Haruta '394 catalyst is used in a hydro-oxidation process.

Iwakura discloses an impregnation method of preparing a catalyst comprising silver, tungsten, and cesium, and optionally, one or more additive metals (gold being mentioned) on a support. The disclosed catalyst is used in the direct oxidation of ethylene with oxygen to form ethylene oxide.

2. Brief Review of the Claimed Invention

The claimed invention pertains to a process of preparing a catalyst composition comprising gold on a titanium-containing support, such as titanium oxide. The process comprises impregnating a gold compound and a reducing agent onto the catalyst support. The claims require that either the reducing agent, or the catalyst support, or both the reducing agent and the catalyst support contain titanium. The catalyst is employed in hydro-oxidation processes, e.g., hydro-oxidizing C3 or higher olefins, e.g. propylene, with oxygen in the presence of hydrogen to yield a corresponding olefin oxide, e.g. propylene oxide.

The claimed impregnation method provides advantages over the co-precipitation method of Haruta '394 in the preparation of supported gold catalysts. As compared with Haruta '394, the claimed method requires less solvent; may employ non-aqueous solvent; does not require controlling pH; can be conducted relatively quickly; provides for improved control over the amount of gold deposited; wastes little gold; and does not require recovery of unused gold from solution.

3. Arguments Against the Rejection

As initial and minimum standards, each reference cited against the application must qualify as prior art under 35 U.S.C. §102 and should be in the field of

Applicant's endeavor, or be reasonably pertinent to the particular problem with which the inventor was concerned. *In re Oetiker*, 24 USPQ2d 14443, 1445 (Fed. Cir. 1992) Based on significant technical differences between Iwakura and the claimed invention, Applicants submit that the skilled artisan would *not* find Iwakura as residing in the field of Applicants' endeavor or reasonably pertinent to the particular problem with which the Applicants were concerned.

Firstly, Iwakura fundamentally pertains to a *silver* catalyst, not a gold catalyst as the Examiner would have. (Iwakura, Abstract; Summary of Invention at Column 1, lines 64-66, continuing onto Column 2, lines 1-44; Claim 1) The Iwakura catalyst requires silver, in a substantial amount from 5 to 50 weight percent, as well as tungsten and cesium. It is taught that, "in some instances" the catalyst may contain one or more additional metals in low concentration (up to 0.01 weight percent). The additional metals read from a "laundry list" of approximately 48 metallic species, including "alkaline metals other than cesium (Group IA), alkaline earth metals (Group IIA), a rare-earth metal, and/or a metal selected from Groups IB, IIB, IIIB, IVA, IVB, VA, VB, and VIA of the Periodic Table as well as tellurium." (Iwakura, Abstract) Iwakura offers no teaching, suggestion, or hint that gold should be selected as the primary element of the catalyst, while the required silver, tungsten, and cesium can each or all be deleted from the catalyst composition. In contrast, the instant claims pertain fundamentally to preparing *gold* catalysts.

Secondly, Iwakura discloses the preparation of a silver catalyst for the *direct oxidation* of ethylene (C2 olefin) with oxygen to form ethylene oxide. In contrast, the claims are concerned with a method of preparing a catalyst for *hydro-oxidation* processes that involve the reaction of a hydrocarbon, such as an olefin, with oxygen in the presence of hydrogen. Hydrogen, not an innocent diluent in hydro-oxidation processes, exhibits a reactive role. Accordingly, the end-use for the catalyst prepared by the invention is fundamentally different from the end-use of Iwakura's catalyst. Iwakura cannot be fairly applied to suggest the preparation of a hydro-oxidation catalyst.

Thirdly, Iwakura relates to a catalyst used in the direct oxidation of *ethylene (C2) with oxygen*; whereas the instant claims relate to preparing a catalyst primarily for the hydro-oxidation of a *C3 or higher olefin*. A history of the art reveals a striking

distinction between directly oxidizing a C2 olefin with oxygen, as compared with directly oxidizing a C3 or higher olefin with oxygen.

> "The preparation of alkylenic oxides starting from olefins and oxygen in the presence of catalysts consisting of mainly of silver and/or silver oxides is known.
>
> In particular, industrial processes for the synthesis of ethylene oxide from ethylene and oxygen or air have been developed and marketed for a long time, whereas up to now interesting results, from the industrial point of view, for the similar synthesis of higher alkylenic oxides, particularly propylene oxide, which is produced indirectly, have not been forthcoming."
>
> US 3,959,316, Column 1, lines 9-19, (copy enclosed)

> "No homogeneous catalyst is known that efficiently catalyzes the epoxidation of olefin with molecular dioxygen. However, this reaction can be achieved with a heterogeneous catalyst, but this process is unique in two respects. First, only ethylene can be epoxidized with high yields, as evidenced by the numerous abortive attempts that have been made to epoxidize higher olefins in this elegant way ....
>
> (2) Why is the epoxidation yield so low for all olefins other than ethylene?
>
> *Cataly. Rev.-Sci. Eng.*, 23 (1 &2), 1981, pp. 128-129 (copy enclosed)

Additional art can be provided to the Examiner consistent with the above teachings.

The art-recognized differences, described hereinabove, place the Iwakura reference at a significant distance from Applicants' field of endeavor. In view of the above, Iwakura does not meet the standard established under *In re Oetiker*, and the reference must be withdrawn.

In the alternative, even if Iwakura is maintained, case law further establishes that when a rejection depends upon a combination of prior art references, there must be some teaching, suggestion, or motivation to combine the references. *Ecolochem, Inc. v. Southern Cal. Edison Co.*, 227 F.3d, 1361, 56 USPQ2D 1065 (FC 2000). There must be some suggestion [for combining prior art references], found either in the references themselves or in the knowledge generally available to one of ordinary skill in the art. *In re Nilssen*, 851 F2d. 1401, 1403, 7 USPQ2d 1500, 1502 (Fed. Cir. 1988) Where does Haruta '394 disclose or suggest that other preparative methods

besides precipitation, and specifically impregnation, can be suitably applied to making a gold-titanium catalyst for hydro-oxidation processes? Applicants can find no such motivation in Haruta '394. Likewise, where does Iwakura teach or suggest that its preparative method is suitable for preparing primarily gold catalysts for hydro-oxidation processes? Applicants can find no such suggestions in Haruta '394 or Iwakura. Accordingly, the required motivation is lacking and the combination must fall.

The Examiner argues that the motivation to combine Haruta '394 and Iwakura is found in Iwakura teaching "a gold catalyst" to make "improved and selective catalysts with long-life." The Examiner twists the teachings in Iwakura to unfairly reject the application. Iwakura clearly pertains in every aspect to a silver catalyst, not a gold catalyst. Moreover, the catalyst of Iwakura is designed for direct oxidation reactions of C2 olefins, not hydro-oxidation processes of C3 and higher olefins. (*Vide supra.*)

The Examiner again regards Applicants' admission that impregnation is a well-known technique in the art as supportive of the rejection. The Examiner fails to comprehend Applicants' rebuttal argument, which was presented through several rounds of prosecution including Applicants' Appeal Brief, filed May 31, 2002 (which the Examiner found to be "persuasive"). Impregnation, like fractional distillation, precipitation, crystallization, and chromatographic separation, is a tool of the chemical trade; but general knowledge of these tools does not imply that each and every tool is applicable to each and every catalyst preparation or chemical process. Indeed, the unpredictability of the catalyst art precludes any generality of that sort. *In re Carleton*, 599 F.2d 1021; 202 U.S.P.Q. 165 (CCPA 1979); *In re Slocombe*, 510 F.2d 1398; 184 U.S.P.Q. 740 (CCPA, 1975); *In re Doumani et al.*, 281 F.2d 215 (CCPA, 1960); 126 USPQ 408. The invention must be taken "as a whole," that is, as a combination of specific elements for a specific outcome. In the instant application, the claimed method requires elements of *impregnation* of a *gold compound* and a *reducing agent* onto a *catalyst support,* wherein the *reducing agent comprises titanium,* or *the catalyst support comprises titanium,* or *both the reducing agent and catalyst support comprise titanium*, for the purpose of preparing a gold-titanium

catalyst for *hydro-oxidation processes.* Applicants' admission offers no substance to the rejection.

Regarding Claim 1, the Examiner argues that Haruta '394 discloses "adding a gold compound with carboxylic acid (see column 3, lines 14-18) and a reducing agent (see column 1, lines 64-65) ..." The Examiner takes words out of context to unfairly reject the application. In fact, Haruta '394 distinguishes between the use of reducing agent and the use of carboxylic acid. Notably, the "(4) Reductant addition method," addressed at Haruta '394, Column 1, lines 64-65, is later described disadvantageously at Column 2:

> " The reductant addition method of 4) has a disadvantage in that when the specific surface area of the metal oxide carrier is small or when the amount of gold loading is increased, for example, the dissolved gold compound is reduced to form metal particles in solution and is not utilized effectively in the deposition of the ultra-fine gold particles on the support oxide and, consequently, the production cost rises."
>
> Haruta '394, Column 2, lines 47-54

The function of carboxylic acid in the co-precipitation method of Haruta '394 is clearly different from that of a reductant:

> "When the pH of the aqueous solution is in the range of 5 to 12 as described above, since the gold compound is incorporated as a hydroxide in the coprecipitate, *virtually no reduction of the gold compound with a carboxylic acid ion occurs*. It is considered that the carboxylic acid ions formed by dissociation in the aqueous solution are adsorbed on the coprecipitate, simultaneously and completely with the gold hydroxide ion $[Au(OH)_4^-]$. *The carboxylic acid ions work as a kind of protective fence for separating the locations at which deposition and precipitation of the gold hydroxide occurs* and thus preventing the gold hydroxide from being agglomerated."
>
> Haruta '394, Column 5, lines 23-37
> *(emphasis added)*

In view of the above, a more accurate interpretation argues that Haruta '394 *teaches away from adding a reductant and that the carboxylic acid is not in this reference a reductant.*

Further in support of Applicants' position, the art "as a whole" teaches away from impregnation in preparing gold catalysts, as evidenced in EP-A1-0,709,360 (of record). Indeed, EP-A1-0,709,360 discloses the impregnation of a titania support with a gold compound, but with *no reductant*, to form an *inoperable* hydro-oxidation catalyst for a C3 olefin. Given that the prior art "taken as a whole" teaches away from reductants and impregnation, it is remarkably unexpected that gold catalysts of good hydro-oxidation activity and high selectivity can be prepared by the combined use of impregnation with a reducing agent. Applicant's discovery significantly simplifies the synthesis of gold catalysts for hydro-oxidation processes and brings the synthesis closer to commercial realization.

At pages 3, 4, and 5 of the Office Action, the Examiner catalogues the alleged teachings of Haruta '394 as relates to Claims 2-9 and 18, 20, 21, 22-23, 24, 25, 26, 27, 29, and 30-32. Applicants have adequately rebutted on the record the rejections of these dependent claims over Haruta '394. Please refer to Applicants' Appeal Brief of May 31, 2002. Iwakura adds nothing substantive to the rejection of the aforementioned dependent claims. Consequently, the arguments will not be repeated again, except briefly as to the following.

Regarding Claims 4 and 5-7, the Examiner argues that Haruta '394 discloses reduction with carboxylic acid. Such is not the case. Haruta '394 describes the function of carboxylic acid as a "protective fence" to keep the gold from agglomerating, which is a different function from that in Applicants' case. Given the scope of the entire art, it would not have been obvious to use a reducing agent, even a carboxylic acid. (*Vide supra.)*

Regarding Claim 20, the Examiner argues that Haruta '394 teaches 0.05 Mol titanium sulfate and an atomic ratio of Au/Ti of 1/19. Implicitly, a Au/TiO_2 mixture having a Au/Ti atomic ratio of 1/19 would have a titanium loading of 53.1 weight percent. The disclosure falls outside the claimed range of greater than about 0.02 to less than about 20 weight percent. Moreover, there is no teaching, suggestion, or hint in Haruta '394 to employ a titanium loading within the claimed range.

With regard to Claims 22, 23, 24, and 27, the Examiner argues that Haruta '394 discloses adding an alkali compound, such as sodium carbonate. Haruta '394

makes no disclosure, suggestion, or hint that the alkali metal ion (e.g., sodium ion) functions as anything other than a source of hydroxide or carbonate for pH adjustment. Note that in every disclosed example the Haruta '394 catalyst is "thoroughly washed," which washing would clearly reduce the sodium concentration. Thus, the intention in Haruta '394 goes against preserving sodium as a promoter to enhance the productivity of the catalyst, as Applicants' claim.

With respect to Claims 10, 13, 14, 19, and 21, the Applicants repeat their rebuttal arguments set forth in previous responses. The Examiner argues that because Haruta '394 discloses "reducing agents comprising carboxylic acids" and use of "titanium both as salts in gold compounds and support material," one skilled in the art would be motivated to select a titanium salt for the reducing agent. As argued earlier on this record, no technical nexus exists between a reducing agent that gives up electrons and a titania support material wherein titanium is in a maximum oxidation state and therefore not capable of giving up electrons. Thus, what the Examiner states as obvious is certainly not so. Moreover, Applicants also assert that Haruta '394 does not actually equate the reducing agents with carboxylic acids.

With regard to Claims 11 and 12, the Examiner argues that Haruta '394 teaches using reducing agents comprising carboxylic acids and salts thereof. Applicants have already addressed the inaccuracy of that statement. (*Vide supra.*) Moreover, Claim 11 requires an organotitanium reducing agent, identified in Applicants' specification as a compound comprising a Ti-C σ–bond or π–bond. Claim 12 requires species of such compounds, namely, alkyltitanium or cyclopentadienyl-titanium compounds, which inherently contain a Ti-C σ-bond or π-bond, respectively. The carboxylic acids and salts of Haruta '394 are characterized by an ionic bond of metal ion to oxide ($M^{+}O^{-}$). No *Ti-C* σ– or π–bond is present in Haruta's disclosure nor are any such compounds suggested. Thus, Claims 11 and 12 are clearly non-obvious.

With regard to Claim 28, the Examiner argues that "Haruta '394 discloses impregnation with solution precipitation;" therefore, any claim to impregnation to incipient wetness is obvious." The Examiner continues to misunderstand Haruta '394 and the technical differences between impregnation and solution precipitation. The Examiner is directed to Applicants' previous responses of record in this file, including

their Appeal Brief of May 31, 2002, for a detailed rebuttal against the notion that Haruta '394 teaches "impregnation with solution precipitation." No more will be said to this point.

In view of the above, it is submitted that Claims 1-9 and 18-32 (and any other claims commented upon by the Examiner in this rejection) are clearly unobvious over Haruta '394 in view of Iwakura. Accordingly, it is respectfully requested that the rejection under 35 U.S.C. §103 be withdrawn.

B. Concerning the Rejection of Claims 10-14 and 16 Under 35 U.S.C. §103(a) Over Haruta '394 in view of Iwakura and Hirose et al.

Claims 10-14 and 16 stand rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Haruta '394 in view of Iwakura further in view of Hirose et al. (US 5,532,030), for the reasons of record. Regarding Claims 10-14, the Examiner continues to validate Hirose et al. on the grounds that this citation discloses a "hydrogenation product catalyst comprising a reducing agent or an acetylacetonate of a titan salt." Regarding Claim 16, the Examiner cites Haruta '394 for disclosing forming a gold/titanium suspension before adding the reducing agent. This rejection is traversed for the following reasons.

Applicants find it inexplicable as to why Hirose et al. is still being cited as a relevant reference. The Examiner is respectfully reminded that the Appeal Brief was found to be "persuasive." In spite of the new rejection, it is time for Hirose et al. to be removed.

The chemical and catalyst arts are well established to be wide-ranging fields and notoriously unpredictable. *In re Carleton*, 599 F.2d 1021; 202 U.S.P.Q. 165 (CCPA 1979); *In re Slocombe*, 510 F.2d 1398; 184 U.S.P.Q. 740 (CCPA, 1975); *In re Doumani et al.*, 281 F.2d 215 (CCPA, 1960); 126 USPQ 408. Accordingly, a citation against the claims must be close or pertinent to the field of the invention. Hirose et al. meets neither criterion. *No disclosure can be found in Haruta '394, Iwakura, or Hirose et al. to motivate the skilled artisan to combine Hirose et al. with the other two references. Hirose et al. relates to* ***polyolefin laminates****, wherein a* ***polyolefin polymer*** *is produced by a* ***ring-opening polymerization reaction of a***

*cycloolefin catalyzed by a **ring-opening polymerization (probably homogeneous) catalyst** comprising titanyl acetylacetonate. In contrast, Haruta '394 relates to preparing gold catalysts for hydro-oxidation processes, while Iwakura relates to ethylene oxidation over a silver catalyst. Regardless that these technologies generally relate to catalysts, each technology is art-recognized as distinctly different over the other two.* Even if the combination of Hirose with Haruta '394 and Iwakura is maintained, there is *no disclosure, suggestion, or hint whatsoever* that titanyl acetylacetonate could be used as a reducing agent in the preparation of a heterogeneous gold-titanium catalyst of suitable activity and selectivity for hydro-oxidizing hydrocarbons, such as, C3 and higher olefins. "Obvious-to-try" is not a criterion for defeating a patent claim.

Claims 11 and 12 are likewise unobvious, because none of Haruta, Iwakura, or Hirose et al. disclose, suggest, or hint at the use of organotitanium compounds containing a Ti-C σ–bond or π–bond as a reducing agent. Since titan acetylacetonate contains a titanium-oxygen ionic bond, Hirose et al. is irrelevant.

As to Claim 16, which requires that gold be deposited prior to the titanium-containing reducing agent, clearly all three cited references are irrelevant.

In view of the above, it is submitted that Claims 10-14 and 16 clearly meet the standards for unobviousness. Accordingly, it is respectfully requested that the rejection under 35 U.S.C. 103 (a) be withdrawn.

C. Concerning Allowable Subject Matter

Applicants gratefully acknowledge the Examiner's finding that Claims 15 and 17 contain allowable subject matter.

D. Conclusions

Applicants believe that the Examiner continues to employ impermissible hindsight to reject the claims in this application. The cited prior art does not motivate Applicants' combination of impregnation with a reducing agent for preparing a gold-titanium catalyst of acceptable activity and excellent selectivity in hydro-oxidation

processes. To the contrary, given the disclosures of EP-0,709,360 and Haruta '394, it can be argued that the art "as a whole" teaches away from impregnation and reduction, which are the very combined directions that Applicants discovered to be successful. Moreover, Applicants' claimed method is superior in several regards to Haruta's precipitation methods *(vide supra)* and much more likely to be adaptable to large-scale commercialization. Without some indication in the cited references that the combined claim elements are desirable, the combination of these elements from non-analogous sources, in a manner that reconstructs the Applicants' invention only with the benefit of hindsight, is insufficient to present a *prima facie* case of obviousness. *Ex parte Dussaud*, 7 USPQ 2nd, 1818, 1820 (PTO Bd. 1988); *In re Grabiak*, 226 USPQ 870, 872 (Fed. Cir. 1985); *In re Fine*, 5 USPQ 2nd 1596, 1598 (Fed. Cir. 1988)

For the record, it is noted that all arguments and evidence submitted previously to the Patent Office in the instant patent application, including Applicants' Response A, Response to the Final Rejection, and Appeal Brief and any submissions therewith, shall be incorporated herein by reference and remain available to the Applicants for any future Appeal.

In view of the above, Applicants maintain that Claims 1 to 32 meet all of the standards for patentability. Moreover, Applicants remain hopeful that the Examiner will find the arguments submitted herein as persuasive as those in the Appeal Brief. A Notice of Allowance is respectfully requested at the Examiner's earliest convenience.

Respectfully submitted,

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